

## Uptake of Trace Elements and PAHs by Fruit and Vegetables from Contaminated Soils

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The aims of this study were to investigate the uptake of seven trace elements and five PAHs in crop plants in order to establish advice regarding consumption of fruit and vegetables grown in soils contaminated by trace elements and PAHs. In a field experiment vegetables were grown in two contaminated soils and in a reference soil, whereas fruits were collected from uncontaminated and contaminated private gardens. The results showed elevated levels of several trace elements and PAHs in the vegetables from contaminated soil. Bioconcentration factors (BCF values) based on dry weight, were below 1, except for those of Cd in lettuce and carrot with peel from uncontaminated soil. In most cases BCF values were decreasing with increasing concentrations in soil. From the heavily contaminated soil BCF values for Pb in lettuce, potato, and carrot with peel were 0.001, 0.002, and 0.05, respectively, and those for benzo[a]pyrene were 0.004, 0.002, and 0.002, respectively. For most metals in most vegetables linear regression showed good correlation between soil and crop concentrations. For PAHs such good correlation was generally not found. The contents of contaminants in fruits were generally low and no correlation with the level of contamination in the soils was found.

### Introduction

Human exposure to contaminants in soil is a matter of general health concern. In urban environments where private gardens are often located in contaminated areas, the exposure of humans to contaminants via consumption of home-grown vegetables and fruit may constitute a potential health risk. Risk assessments which are based on calculations of distribution, transport, and fate in the soil-plant system often include several physicochemical characteristics of the soil (e.g., content of organic matter and pH) in order to predict the availability of the contaminants to crop plants. However, these types of calculations are often erroneously based on the availability of substances in recently spiked soil (1, 2). This seems unrealistic, which has been demonstrated by

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TABLE 1. Toxicological Soil Quality Criteria and Cut-Off Criteria for Soil (Danish EPA 2000)

| substance                | mg/kg dry weight       |                  |
|--------------------------|------------------------|------------------|
|                          | soil quality criterion | cutoff criterion |
| As                       | 20                     | 20               |
| Pb                       | 40                     | 400              |
| Cd                       | 0.5                    | 5                |
| Cr                       | 500                    | 1000             |
| Cu                       | 500                    | 500              |
| Hg                       | 1                      | 3                |
| Ni                       | 30                     | 30               |
| Zn                       | 500                    | 1000             |
| benzo[a]pyrene           | 0.1                    | 1                |
| dibenzo[a,h]anthracene   | 0.1                    | 1                |
| sum of PAHs <sup>a</sup> | 1.5                    | 15               |

<sup>a</sup> Sum of the PAHs fluoranthene, benzo[b+j+k]fluoranthene, benzo[a]pyrene, dibenzo[a,h]anthracene, and indeno[1,2,3-cd]pyrene.

extensive research on the behavior of heavy metals and organic xenobiotics in soil, including the availability of the substances to percolating water and live organisms (3, 4). These studies showed that the availability of most substances in soil is considerably reduced by aging, due to the formation of strong bonds to soil particles and/or incorporation into resistant secondary minerals of low bioavailability (e.g., iron oxides). Therefore, the uptake rate of contaminants by crop plants should be determined in experiments conducted in the genuine contaminated soil. On the basis of toxicological assessments of a number of substances, the Danish EPA (5) has defined soil quality criteria (no restrictions on land use) and cutoff criteria (any contact with the soil should be omitted) as shown in Table 1. In the interval between these criteria (the advisory interval), advice will be given regarding the use of the soil in order to avoid unacceptable human exposure with respect to intake of home-grown crops (5). In Denmark, it was decided to investigate whether the general advice to the public (not to eat any crop grown in soil which is contaminated above the soil quality criterion) could be revised if realistic experiments were used as a basis of the risk assessments.

The aim of the present investigation was to establish appropriate and detailed advice regarding consumption of fruit and vegetables grown on contaminated land. Preliminary sampling of limited numbers of vegetables and fruits (including nuts and berries) in Copenhagen (6) indicated that the level of inorganic contaminants in the crops was low. To study the plant uptake of elements and organic substances from contaminated soil, field experiments included cultivation of vegetables grown in soils containing trace elements and PAHs and collection of samples of fruits from contaminated gardens. The implication to human health was evaluated by estimation of the intake of the contaminants via the total diet, including contaminated vegetables and fruit. This paper describes the experimental part of the investigations.

### Materials and Methods

**Vegetable Cultivation Experiment.** Vegetables were grown in soils from three sites: an uncontaminated agricultural field ("uncontaminated soil"), a site in Copenhagen contaminated from diffuse sources ("medium contaminated"), and a heavily contaminated soil from a former landfill near Copenhagen ("heavily contaminated"). The medium contaminated soil was selected in order to achieve concentrations

TABLE 2. Vegetable Crops Used for Analysis of Trace Elements and/or PAHs

| crop                                     | analyzed for trace elements | analyzed for PAHs |
|--|-----------------------------|-------------------|
| lettuce ( <i>Lactuca sativa</i> )        | +                           | +                 |
| potato ( <i>Solanum tuberosum</i> )      | +                           | +                 |
| carrot ( <i>Daucus carota</i> )          | +                           | +                 |
| radish ( <i>Raphanus sativus</i> )       | +                           | -                 |
| green bean ( <i>Phaseolus vulgaris</i> ) | +                           | -                 |
| squash ( <i>Cucurbita pepo</i> )         | -                           | +                 |

of most contaminants within the advisory interval (5). The original soil from each of the three sites was arranged in a common location close to Copenhagen in piles of approximately 12 m x 12 m and a depth of 0.5 m. The soils were fertilized to equal levels of N, P, and K and cultivated with a rotary cultivator as in frequent garden practice.

In each of the three soils, 6 crops were grown in 10 replicates in a randomized complete block design with 10 blocks and 6 plots in each soil. Before sowing, soil samples were taken in each plot by combination of five drills (0–50 cm) taken from the corners and from the center of a 1 m x 1 m square. From the uncontaminated soil, only one sample was taken per block. The crops were sown in rows which were separated by a distance of 30 cm and starting at a distance of 30 cm from the edge of the plot. The crop species were selected to represent a worst case uptake of metals and PAHs as based on literature information and also to represent a significant mass fraction of the Danes' diet. The selected crops (Table 2) were grown to maturity before harvest and each plot was harvested completely. The experiment was started in June and the crops were harvested in October 1999. From the experimental plots, 5 samples of each crop and samples of the corresponding soil were analyzed for trace elements and/or PAHs. Additionally, analyses of soil parameters were carried out. Measurements of plant data such as growth rate, yield, or height were not made.

**Sampling of Fruits.** Samples for trace element analysis were collected in existing allotment gardens in the Copenhagen area. One allotment garden area which was only slightly contaminated was chosen as the local reference area whereas the eight other allotment areas were "medium" to "heavily" contaminated with trace elements. From one of the medium-heavily contaminated allotment areas (Kälvbod) the numbers of samples of individual fruits was higher than those from the other contaminated areas, and data from this area were treated separately. For PAH analyses, samples of soil as well as of fruits were collected in Skagen (Northern Jutland) in PAH-contaminated private gardens which were former drying grounds for tar-impregnated fishing nets. For comparison, fruits were sampled from gardens expected to be uncontaminated. The selection of species of fruits was based on criteria regarding consumption rate, taxonomy (to

represent different families) exposure conditions (surface-to-volume ratio, duration of growth period), and their occurrence in the gardens. The available plant species in Copenhagen were different from those in Skagen which is reflected in Table 3. Hazelnuts were included because analysis (unpublished data) of market nuts at the Danish Veterinary and Food Administration had shown very high concentration levels of trace elements.

**Pretreatment of Samples.** All samples were thoroughly washed to remove all visible soil particles before analysis. The samples of carrots and potatoes were split into two pieces and one half was peeled. After pretreatment, the samples from the same site were cut into small pieces which were mixed before sub-samples were taken at random from the total sample for the final chemical analysis. To reduce the number of analyses and still have as many gardens as possible represented, composite samples of fruits were prepared by mixing of sub-samples from several gardens with comparable levels of soil contamination.

#### Chemical and Physical Analysis of the Crops and Soils.

The dry matter content of soil and crop samples was determined, and soil and crop samples were analyzed for the trace elements arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn). Soils were analyzed for the following PAHs: naphthalene, acenaphthylene (Ace), acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene (Flu), pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene (BbF), benzo[j+k]fluoranthene (BjkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthracene (DahA), and benzo[ghi]perylene. The additional soil parameters comprised total carbon, organic carbon, CaCO<sub>3</sub>, texture (clay, two silt and five sand fractions), sodium, potassium, calcium, and magnesium ions, as well as the cation exchange capacity and pH. The PAHs analyzed for in plants included Ace, Flu, BbF, BkF, BaP, and IcdP. On the basis of bids, the Danish EPA selected five different laboratories in Denmark to carry out the analyses. Soil parameters were analyzed at The Danish Institute of Agricultural Sciences, Research Centre Foulum; soil metals and PAHs were analyzed at AnalyCen Analytical Laboratory; metals in vegetables were analyzed at The Copenhagen Food Control Laboratory; metals in fruits were analyzed at DHI Water & Environment; and PAHs in all plant samples were analyzed at Risø National Laboratory.

**Determination of Trace Elements.** Soil samples were extracted by nitric acid at 120 °C. Arsenic was analyzed by hydride generation-AAS and the remaining metals were analyzed by ICP-AES. In all analytical series, a sample of certified reference material (CRM), a blank, and a determination in duplicate were included for assurance of analytical accuracy. The results of the analysis of the CRM showed deviations from the certified value of generally 0–10% for all elements.

TABLE 3. Numbers of Samples of Fruits Collected for Analysis of Trace Elements and PAHs

|                                      | trace elements (Copenhagen) |                | PAHs (Skagen) |                |
|--------------------------------------|-----------------------------|----------------|---------------|----------------|
|                                      | contaminated                | reference area | contaminated  | reference area |
| pear ( <i>Pyrus communis</i> )       | 57                          | 8              | 4             | 10             |
| apple ( <i>Malus domestica</i> )     | 0                           | 0              | 16            | 9              |
| plum ( <i>Prunus domestica</i> )     | 43                          | 11             | 10            | 10             |
| elderberry ( <i>Sambucus nigra</i> ) | 44                          | 8              | 0             | 0              |
| blackberry ( <i>Rosa pilicatus</i> ) | 31                          | 7              | 0             | 0              |
| other berries <sup>a</sup>           | 20                          | 2              | 8             | 9              |
| hip ( <i>Rosa rugosa</i> )           | 0                           | 0              | 11            | 11             |
| hazelnut ( <i>Corylus avellana</i> ) | 13                          | 7              | 0             | 0              |

<sup>a</sup> Other berries included the following: for trace elements, gooseberry (*Ribes uva-crispa*), blackcurrant (*Ribes nigrum*), and redcurrant (*Ribes rubrum*); for PAHs, gooseberry.

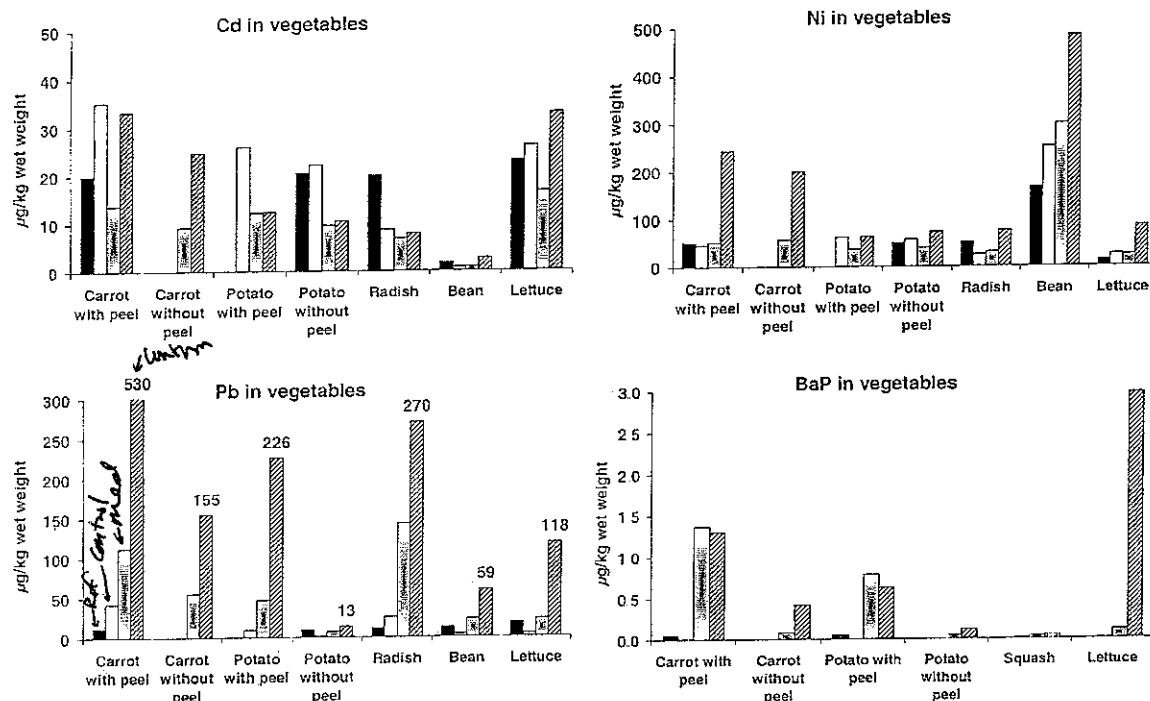


FIGURE 1. Cd, Ni, Pb, and BaP in vegetables from three experimental soils as well as background values. Open bars, reference soil; shaded bars, medium contaminated soil; hatched bars, heavily contaminated soil; black bars, reference values (i.e., metals in Danish market crops (7) and BaP mean background level, based on literature as described in the text) Carrot without peel from the reference soil was not analyzed.

Vegetables were digested under pressure with nitric acid at 160 °C, and the contents of As, Cd, Pb, Ni, and Cr were analyzed by graphite furnace-AAS with Zeeman background correction, and Cu and Zn were analyzed by ICP-AES. The CRM Standard Reference Material 1572, citrus leaves (U.S. National Institute of Standards and Technology; NIST) was included in every batch of analyses (up to 12 samples) together with a blank and a determination in duplicate.

Fruits, berries, and nuts were digested using nitric acid in a microwave oven. Ni was analyzed by the graphite furnace AAS technique, whereas high-resolution ICP-MS was used for analysis of As, Cd, Cr, Cu, Pb, and Zn. Reference materials analyzed in parallel with the samples controlled the accuracy of the analyses. The CRMs used for the fruit analysis were tomato leaves SRM 1573au (NIST) and sea lettuce BCR 279 (for Pb) (EU Community Bureau of Reference), which were analyzed in parallel with the samples. For each fruit type, determinations in duplicate were also made. For both vegetables and fruits, the results of the analyses were within the accept intervals.

**Determination of PAHs.** Soil samples were shaken in sodium pyrophosphate and toluene for 16 h and analyzed by GC-MSD. Samples of vegetables and fruits were extracted as follows: 10 g of homogenized plant material was extracted with hexane/acetone (1:1) in Soxhlet (a combination of Soxhlet and microwave technique). The extract was transferred to centrifuge glasses and the plant material was rinsed three times with hexane/acetone (1:1). The extract was centrifuged subsequently cleaned in a SPE/NH<sub>2</sub>-SAX-column (solid phase extraction) and finally evaporated. The analyses were carried out by HPLC with fluorescence detection and verification of selected samples by GC/MS. The samples were analyzed in batches with up to 10 samples plus one blank, one reference sample, and a duplicate determination. CRMs for PAHs in plant tissue were not available, and internal standards were produced by addition

of certified reference solutions (Dr. Ehrenstorfer) to subsamples of the plant material followed by recording of recovery. Recovery was between 80% and 95% for all substances (lowest for Ace) and the coefficient of variation was in the range of 11–21%.

## Results

**Vegetable Field Experiment.** The lettuce and carrot plants were poorly developed in many plots, especially in those containing heavily contaminated soil. This could be due to toxic effects of the contaminants, but even if the experiment was watered regularly, drought cannot be excluded as a cause. Supplementary lettuce and carrot were sown, but the resulting plants were very small at harvest. The small carrots were irregular and difficult to peel, which may have resulted in remnants of peel in the peeled samples.

**Analyses of Soil.** The results of the soil parameter analyses showed that the soil types were different. The organic matter content was approximately 3% in both the uncontaminated soil and the medium contaminated soil, but the heavily contaminated soil (from a former landfill) contained 19% organic matter. The origin of this was probably organic waste. The main difference between the uncontaminated and the medium contaminated soils was that the uncontaminated soil was more clayey than the medium contaminated soil (levels of clay-silt were 31% and 16%, respectively). The results of the trace element and PAH analyses of the soils are presented in detail in the Supporting Information (SI Table 1). Despite a large variation of concentrations measured, the mean values from the three soils were significantly different ( $P < 0.05$  by ANOVA). In the uncontaminated soil, the concentrations of all substances were below the soil quality criteria as defined by the Danish EPA (5), with the exception of BaP in one sample. In the medium contaminated soil, the concentrations of Cd, Pb, BaP, and DahA were close to the advisory intervals (5), whereas the concentrations of the

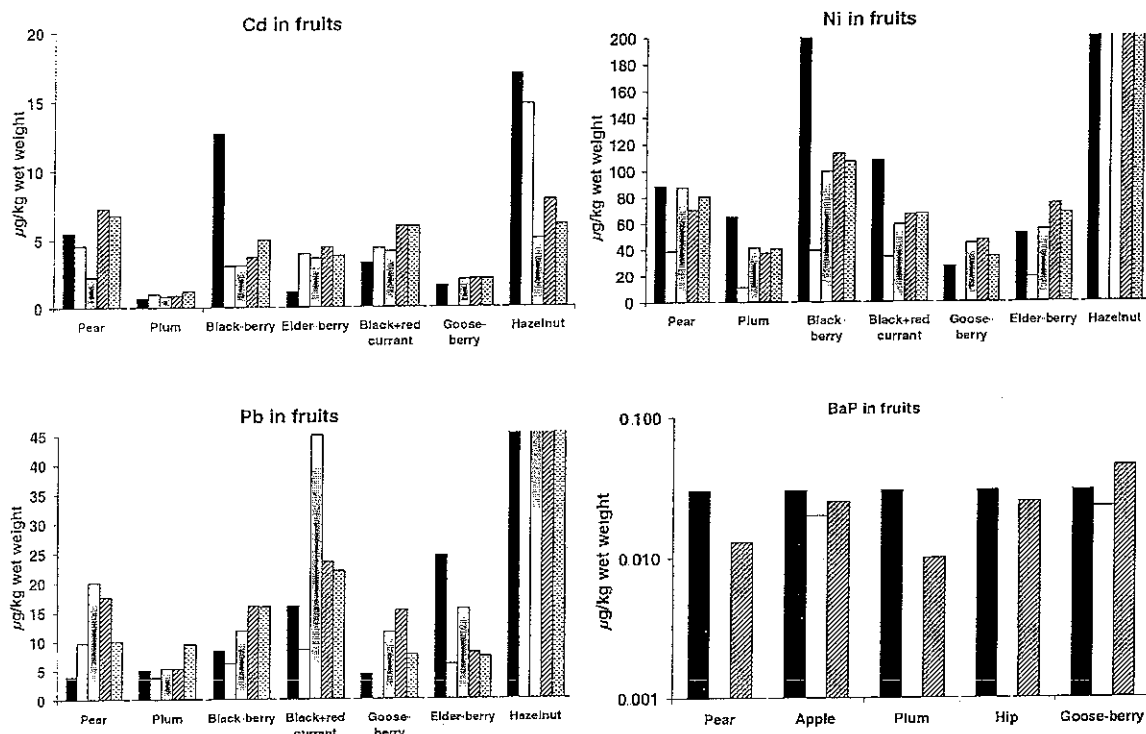


FIGURE 2 Cd, Ni, Pb, and BaP in fruits from reference areas and contaminated areas as well as background values. Open bars, reference areas; shaded bars, medium contaminated soil; hatched bars, heavily contaminated soil; dotted bars, Kalvebod area; black bars, background values (i.e., trace elements in Danish market fruits (7) and BaP mean background level, based on literature as described in the text). Values for Ni and Pb in hazelnut not shown.

remaining substances were below the soil quality criteria. In the heavily contaminated soil, the mean concentrations of all substances were above the cutoff criteria. The mean concentrations (all dry-weight based) of Pb were 20, 130 and 1000 mg/kg and of BaP were 0.1, 2 and 15 mg/kg in the three soils, respectively, but the spread was large. To illustrate this, the highest concentrations of the two substances in the heavily contaminated soil were 11 000 and 160 mg/kg respectively. The range of Cd concentrations in the three soils was narrower, with means of 0.33, 0.56 and 2.4 mg/kg respectively.

**Trace Elements in Vegetables.** The concentrations (all wet-weight based) of trace elements Cd, Ni and Pb in vegetables are summarized in Figure 1 and results for all trace elements are given in detail in SI Table 2. For comparison, the results of available measurements on market crops from the Danish monitoring system (7) are included. The concentrations of most trace elements in the crops increased with the degree of contamination of the soils (Figure 1 and SI Table 2) except Cd, and in some crops Ni. The concentration of As was generally below the detection limit (12 µg/kg) except in the three root crops with peel. The concentrations of Pb in all crops from the two contaminated soils were considerably higher than those of market vegetables, while the picture was less clear for Cd and Ni.

**PAHs in Vegetables.** The results (Figure 1 (BaP) and SI Table 3) showed that the content of Ace was below the detection limit (0.5 µg/kg wet weight) in all crops and that IcdP was found only in the two root crops with peel from the two contaminated soils and in carrot without peel from the most contaminated soil. In most crops the concentrations of most of the PAHs were significantly higher in crops from contaminated soils than in those from the uncontaminated soil. However, for all PAHs measured, there were no significant differences between concentrations in crops from

the two contaminated soils, except for potatoes and carrot without peel. Peeling of the potato and carrot samples reduced the concentrations of BaP and IcdP considerably. Reported concentrations in root crops range from 0.01 to 0.1 µg/kg wet weight (indicated in Figure 1) while the range of levels in leafy vegetables is very wide (0.2–48 µg/kg wet weight) and gives no basis for selection of a background level (8–11).

**Sampling of Fruits.** The levels of trace element concentrations (all dry weight based) in the soils from allotment areas used for the sampling of fruits are summarized in SI Table 4. In the reference area, the concentrations were below those of the advisory interval except for Pb for which the mean concentration was 82 mg/kg dry weight. In the medium and heavily contaminated areas, the ranges of Pb were 432–655 and 853–1248 mg/kg, respectively. Cd concentrations were within the advisory interval (0.5–5 mg/kg) in all areas, whereas the Ni concentrations were above the cutoff criterion of 30 mg/kg in all contaminated areas (maximum at 58 mg/kg).

The soil PAH concentrations (all dry weight based) in the contaminated areas are summarized in SI Table 5. The concentrations in the reference gardens were not measured but were expected to be below the soil quality criteria, which are defined for BaP (0.1 mg/kg), DahA (0.1 mg/kg), and the sum of PAHs (1.5 mg/kg) by the Danish EPA (5). In the contaminated gardens the concentrations of BaP ranged from 0.18 to 33 mg/kg.

**Trace Elements in Fruits.** The results of the trace element analyses of fruits are summarized in Figure 2 for Cd, Ni, and Pb, and results for all trace elements are provided in SI Table 6. For comparison, the results of available data for market crops from the Danish monitoring system (7) are included. The levels of trace elements in fruits from the investigation and in market fruit were highly variable, and in several cases

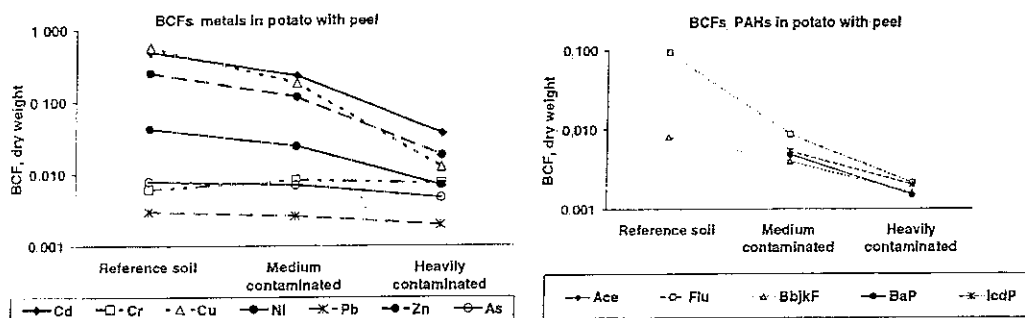


FIGURE 3 BCF values for trace elements and PAHs for potato with peel from three experimental soils.

the mean concentrations recorded from market fruit were above the levels measured in the investigation. The levels in hazelnut were up to 1 order of magnitude higher than the levels in fruits with means ranging from 6 to 18 400–1800 and 100–450  $\mu\text{g}/\text{kg}$  of Cd, Ni and Pb respectively. Among these the market nuts (of unknown origin) contained the highest levels of Cd and Ni.

**PAHs in Fruits.** The results of the analyses of PAHs in fruits (Figure 2 (BaP) and SI Table 7 (all PAHs)) were dominated by results below the detection limit. Ace was not detected in any sample because of the relatively high detection limit and losses were possibly caused by evaporation from the samples during the extraction process. Traces of IcdP were detected in a few samples of apple and gooseberry. The mean reference level of BaP of 0.03  $\mu\text{g}/\text{kg}$  wet weight (Figure 2) is based on literature background concentrations of BaP in fruits showing a range of 0.02–0.04  $\mu\text{g}/\text{kg}$  wet weight (8). The only crop exceeding this interval was gooseberry with a mean concentration of 0.045  $\mu\text{g}/\text{kg}$  wet weight in samples from contaminated gardens.

## Discussion

**Vegetables. Concentrations.** For several crops and trace elements the concentrations (all based on wet weight) of trace elements in vegetables grown in contaminated soil were higher than those of market crops (7) (Figure 1 (Cd, Ni, Pb) and SI Table 2 (all trace elements)). Lead was the only trace element for which this was valid for all crops and the levels of Pb measured in crops from the contaminated soils were very high compared to those of market vegetables. The concentrations of BaP measured in the root crops with peel from the contaminated soils (up to 1.4  $\mu\text{g}/\text{kg}$ ) were well above the background level established from literature data. For a generally applicable assessment of the correlation between the concentrations in the soil and the concentrations in crops the bioconcentration factors (BCF values) were calculated.

**BCF Values.** BCF values were calculated as the ratio between the concentrations in plants and soil (all based on dry weight) for each plot separately. The means of the calculated BCF values for each crop and soil are presented in SI Tables 8 (trace elements) and 9 (PAHs). As an example BCF values for potato with peel of trace elements and PAHs are shown in Figure 3.

The BCF values were below 1 except for values for Cd in lettuce and carrot with peel from the uncontaminated soil. From the heavily contaminated soil BCF values for Pb in lettuce, potato and carrot with peel were 0.001, 0.002 and 0.05, respectively, and those for benzo[a]pyrene were 0.004, 0.002 and 0.002, respectively. This means that the concentrations in soil, which may remain on the vegetables after preparation in the kitchen, can be several orders of magnitude higher than the concentrations in the vegetables themselves. In most cases BCF values were decreasing with increasing concentrations in the soil, especially for the more mobile substances (Cd, Ni, Zn, Ace, and Flu) for which the highest

BCF values were found. In the heavily contaminated soil the high organic matter content (19%) may have reduced the availability of several substances. For some metals a similar inverse relationship has been recorded in other investigations in terrestrial environments, which was discussed and referred to as a "saturation effect" by Dudka & Miller (1). Therefore the level of contamination may be important in order to compare BCF values in plants grown in different soils (experiments).

Dudka & Miller (1) reported BCF values for Cd, Pb and Zn in potato with peel from soil contaminated with flue dust at levels 5–10 times the concentrations in the heavily contaminated soil of the present investigation. These were in the same order of magnitude as those found here indicating that the availability of the trace elements in the flue-dust contaminated soil was higher than that in the highly contaminated former landfill soil. Larsen et al. (12) reported BCF values for Cr and As in potato and carrot with peel from an experiment in soil contaminated by atmospheric deposition. The levels of BCF for As were comparable to that of the heavily contaminated soil of this investigation, whereas the level of BCF for Cr was lower by a factor of 4. The results therefore point to the fact that the origin – and possibly the age, which is unknown – of the contamination are important for the availability of elements in soil.

For the PAHs, similar comparisons can be made to the results of German experiments (13, 14). In these experiments, the importance of direct exposure of leaf crops to soil was investigated by covering the soil with uncontaminated sand. It was demonstrated that the main route of exposure to PAHs for leaf crops was via direct contact to soil on the leaves. The results of the German investigation gave BCF values for carrot with peel similar to those of the present investigation. The BCF values reported from the German study for lettuce, which was directly exposed to soil, exceeded those of the present investigation with a factor of about 10. This could be due to differences in the varieties of lettuce used in the two experiments, because the variety used by Delschen et al. (13) was very open, allowing soil to reach all leaves, whereas that of this investigation had tight heads.

**Correlation with Concentration in Soil.** To use the results of the experiment for a generally applicable exposure assessment, the relationship between concentrations of individual substances in soils and in crops was examined by regression analysis. For PAHs (i.e., Flu, Bb/kF, and BaP),  $R^2$  values from linear regressions were below 0.2 and the  $P$  values were far above 0.1 for all PAHs in all crops except lettuce (and Flu in potatoes). Even though logarithmic regression improved this, still no  $R^2$  value was above 0.6. Therefore, regressions were not considered to be an appropriate tool for predicting concentrations of PAHs in vegetables at advisory interval soil concentrations.

For the trace elements correlations were higher, and results of the linear regression analysis are summarized for all trace elements in SI Table 10 and illustrated in Figure 4.

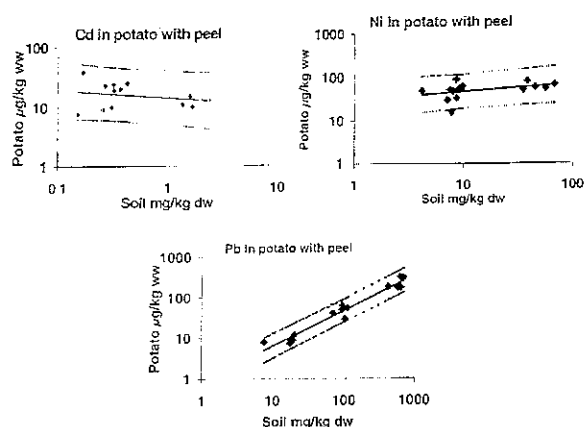


FIGURE 4 Linear regressions for Cd, Ni, and Pb in potato from three experimental soils.

for Cd, Ni, and Pb in potato.  $R^2$  values were relatively high ( $>0.7-0.96$ ) for Cr, Cu, Pb, and Zn in almost all crops, whereas correlation was poor for Ni ( $0.2-0.9$ ) and As ( $0.03-0.65$ ) and very low for Cd ( $0.006-0.4$ ).

The lack of correlation for Cd could be due to increased availability of Cd in the uncontaminated soil because phosphorus fertilizers used on agricultural soil are often contaminated with this metal (15) and consequently Cd has not been aged for long in agricultural soil.

The importance of the soil properties for the availability of the substances was examined by means of partial least squares analysis (16). However, the organic carbon content of the heavily contaminated soil was very high (19%) and this seems to have overshadowed the influence of other possible parameters such as pH, CEC, and clay content, which only varied to a limited degree between the three soils. Even though some trace elements in some crops showed reduced availability with increasing pH or increasing CEC, the only common trend was reduced availability with increasing carbon content.

In summary, all trace elements except Cd were found in higher concentrations in all types of vegetables from the contaminated soils than from the reference soil. Furthermore, peeling of root crops (potato and carrot) removed most of the contamination, indicating that the contamination was caused by uptake due to direct soil contact and not by translocation. For the health risk assessments, expected concentrations of trace elements in vegetables at soil concentrations equivalent to the upper limit of the advisory interval (the cutoff criterion) could be predicted by means of the equations from the regression analysis. For PAHs, this option was not considered applicable and the risk assessment was based on measured concentrations in vegetables from the medium-contaminated soil.

**Fruits.** The levels of the trace elements and PAHs measured in fruits were generally low compared to those of the vegetables.

**Trace Elements.** The concentrations of trace elements in fruits (Figure 2 and SI Table 6) from contaminated areas were compared to the concentrations in crops from the reference area and to those in market crops (7). The level of Pb in pear from all gardens (including the reference area) was significantly higher ( $P < 0.05$  in Tukey's  $t$ -test) than in market fruit. As Pb is not being translocated readily in plants, it could be suggested that the Pb found in pears originated from atmospheric deposition. However, if this was the case, the fruits of plum and especially elderberry, which have large surface-to-volume ratios, should also show elevated levels of Pb, which was not the case. In berries from bushes, the levels of Pb were slightly higher in berries from all gardens

than those in market berries. The concentrations of Ni and Cd in pear, plum, and elderberry were generally at the same level as those of market fruits except that the level of Cd in elderberry was higher from all gardens than in market elderberry. In the other berries, there was no general trend to elevated levels of Ni and Cd. Even though some examples of elevated levels of trace elements in fruits from contaminated areas were found, there were no correlations between increasing levels of Pb, Cd, or Ni in the soil of the allotment areas and the levels in fruits.

**PAHs.** Traces of IcdP were detected in a few samples of apple and gooseberry. This substance is known to be bound strongly to soil and is not expected to be translocated in plants. In the vegetables, it was only measured in samples of root crops with peel from contaminated soil. The IcdP found in gooseberry could be due to sorption from splashes of soil on the berries during growth. The contents in apple may originate from atmospheric sources but no information is available regarding other possible sources of PAH contamination in the gardens—e.g., presence of woodburning stoves or hobby smoking chambers. There was no correlation between the concentrations measured in fruits and the level of contamination of the soil in the gardens—even though there was a tendency to lower levels in fruits from uncontaminated gardens. For BaP, the levels found in fruits from contaminated gardens were  $<0.018-0.045 \mu\text{g/kg}$  wet weight, which is similar to the literature-based reference level of  $0.02-0.04 \mu\text{g/kg}$  wet weight.

In summary, levels of trace elements and PAHs in fruits varied between samples from reference areas and contaminated areas, but no correlations were found between concentrations of contaminants in the soil and concentrations in the fruits. However, the possibility of contamination of berries, which were growing close to the soil surface through direct uptake from soil deposited on the surface, could not be excluded.

#### Acknowledgments

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#### Supporting Information Available

Soil and crop concentrations of all contaminants, BCF values for elements and PAHs in vegetables, and parameters from linear regression analysis of correlation between soil and crop concentrations of elements for vegetables. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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